Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

Supplementary information for *"Magnetic properties and quench dynamics of two interacting ultracold molecules in a trap"*

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I. DERIVATION OF THE SIZE OF THE MOLECULAR CLOUD IN THE USED BASIS

The wave function of the studied molecular system is following:

$$|\Psi_k\rangle = \sum_{\substack{n,J,M,j_1,j_2,\\ S,M_S,s_1,s_2}} C^k_{n,J,M,j_1,j_2,S,M_S,s_1,s_2} |n\rangle |J,M,j_1,j_2\rangle |S,M_S,s_1,s_2\rangle \,,$$

where

$$\begin{aligned} |n\rangle &= \frac{1}{\sqrt{2^n n!}}\pi^{-1/4}\exp\left\{\frac{-z^2}{2}H_n(z)\right\}, \\ |J,M,j_1,j_2\rangle &= \sum_{m_1,m_2} \langle j_1,m_1,j_2,m_2|J,M\rangle |j_1,m_1\rangle |j_2,m_2\rangle \,, \\ |S,M_S,s_1,s_2\rangle &= \sum_{m_{s_1},m_{s_2}} \langle s_1,m_{s_1},s_2,m_{s_2}|S,M_S\rangle |s_1,m_{s_1}\rangle |s_2,m_{s_2}\rangle \,. \end{aligned}$$

where H_n are the Hermite polynomials, $\langle j_1,m_1,j_2,m_2|J,M\rangle$ and $\langle s_1,m_{s_1},s_2,m_{s_2}|S,M_S\rangle$ are the Clebsch-Gordan coefficients, while $|j_i,m_i\rangle$ and $|s_i,m_{s_i}\rangle$ are the eigenfunctions of the rotational and spin angular momenta of the molecule i. The size of the molecular cloud is then (with $\beta \equiv J,M,j_1,j_2,S,M_S,s_1,s_2$):

$$\langle \hat{r}^2\rangle = \langle\Psi_k|\,\hat{r}^2\,|\Psi_k\rangle = \sum_{n,n',\beta} C^k_{n,\beta}C^k_{n',\beta}\,\langle n|\,\hat{r}^2\,|n'\rangle = \sum_{n,n',\beta}\frac{C^k_{n,\beta}C^k_{n',\beta}}{\sqrt{2^{n+n'}n!n'!}}\pi^{-\frac{1}{2}}\int_0^\infty dr\, r^2 e^{-r^2}H_n(r)H_{n'}(r)$$

$$\begin{aligned} \int_0^\infty dr\, r^2 e^{-r^2}H_n(r)H_{n'}(r) &\overset{①}{=} n!n'!\sum_{N=0}^{\min n,n'}\frac{2^N}{(n-N)!(n'-N)!N!}\int_0^\infty dr\, r^2 e^{-r^2}H_{n+n'-2N}(r) \\ &\overset{②}{=} n!n'!\sum_{N=0}^{\min n,n'}\frac{2^N(n+n'-2N)!}{(n-N)!(n'-N)!N!}\sum_{k=0}^{\mathrm{floor}(\frac{n+n'}{2}-N)}\frac{(-1)^k 2^{n+n'-2N-2k-1}\Gamma(\frac{n+n'-2N+3}{2}-k)}{k!(n+n'-2N-2k)!} \end{aligned}$$

Hermite polynomials' properties used in calculations:

① $H_m(z)H_n(z) = m!n!\sum_{N=0}^{\min m,n}\frac{2^N H_{m+n-2N}(z)}{(m-N)!(n-N)!N!}$ [1],

② $\int_0^\infty t^2 e^{-t^2}H_n(t)dt = n!\sum_{k=0}^{\mathrm{floor}(\frac{n}{2})}\frac{(-1)^k 2^{n-2k-1}\Gamma(\frac{n+3}{2}-k)}{k!(n-2k)!}$ [2].

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II. SPIN-ROTATION COUPLING MATRIX ELEMENTS IN THE USED BASIS

Here, we provide matrix elements of the spin-rotation component of the Hamiltonian given by Eq. (1) and (2) in the computation basis of $|n, J, M_J, j_1, j_2, S, M_S, s_1, s_2\rangle \equiv |n\rangle|J, M_J, j_1, j_2\rangle|S, M_S, s_1, s_2\rangle$ as described in Sec. 2 of the main text.

$$
\begin{gathered}
\langle n, J, M_J, j_1, j_2, S, M_S, s_1, s_2|\hat{H}_{\text{spin-rot}}|n', J', M'_J, j'_1, j'_2, S', M'_S, s'_1, s'_2\rangle = \\
\delta_{nn'}\delta_{M_{\text{tot}},M'_{\text{tot}}}\delta_{j_1j'_1}\delta_{j_2j'_2}\delta_{s_1s'_1}\delta_{s_2s'_2} \\
\times \sum_{m_1=-j_1}^{j_1}\sum_{m_2=-j_2}^{j_2}\sum_{m'_1=-j_1}^{j_1}\sum_{m'_2=-j_2}^{j_2} \langle j_1m'_1j_2m'_2|JM_J\rangle\,\langle j_1m'_1j_2m'_2|J'M'_J\rangle \\
\times \sum_{m_{s_1}=-s_1}^{s_1}\sum_{m_{s_2}=-s_2}^{s_2}\sum_{m'_{s_1}=-s_1}^{s_1}\sum_{m'_{s_2}=-s_2}^{s_2} \left\langle s_1m'_{s_1}j_2m'_2\middle|SM_S\right\rangle\left\langle s_1m'_{s_1}s_2m'_{s_2}\middle|S'M'_S\right\rangle \\
\times\Big(\delta_{m_1m'_1}\delta_{m_2m'_2}\delta_{m_{s_1}m'_{s_1}}\delta_{m_{s_2}m'_{s_2}}\gamma(m_1m_{s_1}+m_2m_{s_2}) \\
+\frac{\gamma}{2}\Big(\delta_{m_{s_1}+1,m'_{s_1}}\delta_{m_{s_2},m'_{s_2}}\delta_{m_1-1,m'_1}\delta_{m_2m'_2}+\delta_{m_{s_1},m'_{s_1}}\delta_{m_{s_2}+1,m'_{s_2}}\delta_{m_1m'_1}\delta_{m_2-1,m'_2} \\
+\delta_{m_{s_1}-1,m'_{s_1}}\delta_{m_{s_2},m'_{s_2}}\delta_{m_1+1,m'_1}\delta_{m_2,m'_2}+\delta_{m_{s_1},m'_{s_1}}\delta_{m_{s_2}-1,m'_{s_2}}\delta_{m_1m'_1}\delta_{m_2+1,m'_2}\Big)\Big)\ ,
\end{gathered}
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[1] G. N. Watson, J. London Math. Soc. **s1-13**, 29 (1938).
[2] Wolfram Research, Inc., "The mathematical functions site," (2020), function ID: 05.01.21.0009.01.